Variation in Lake Water Chemistry and Spatial Scale

Analysis of the Swedish National Lake Monitoring Programme

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Abstract

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Lake water chemistry generally reflects the ecological, geological and climatic processes in and around a lake. Spatial variability both within and among lakes affects our interpretation of lake monitoring data. This thesis addresses the importance of scale, from small (variability within a lake and among neighboring lakes) to large (ecoregions, EMEP-grid and parts of the country) on selected chemical metrics using data from the Swedish lake monitoring program. Different approaches are used when studying the spatial variability of lake water chemistry, *e.g.* partial least squares regression (PLS) and variogram modeling. The synchronous variation of lake water chemistry is also investigated.

Paper I addressed the question of how representative a mid-lake surface sample was of in-lake conditions. Where a sample was taken within a lake was found to affect the sample's representativity of lake water surface chemistry. As a mid-lake sample was shown to be more representative than a random sample, my conclusion was that a mid-lake sample can be considered representative of average surface water chemistry.

Partition lakes regarding lake water chemistry is an important issue for interpreting lake monitoring data. In *Paper II* lakes were classified into six ecoregions. The study showed that ecoregional classification of lake water chemistry only worked in the southern part of Sweden, where there was sufficient variation within the ecoregions. Both *Paper III* and *IV* indicated that water-chemical data should be interpreted at a regional scale (*e.g.* EMEP-scale). In *Paper III* I showed that there was a spatial dependence in lake water chemistry; Ca^* and Mg^* concentrations in lakes close to each other were more similar than in lakes at further distances, but the distance for dependence can vary within the country. *Paper IV* showed that lakes seemed to have high rate of synchronous variation at least for the variables SO_4^*, Ca^*, AbsF and ANC.

Further knowledge of the connection between variation in lake water chemistry and *e.g.* weathering, soil chemistry, soil type, distribution of mires, land use, precipitation, deposition and runoff are needed. An interesting task would be to develop a method to classify the country into regions regarding lake water chemistry together with experts in geology and hydrology.

Key words: regionalization, predictive models, pH, total phosphorus, catchment characteristics, geostatistics, liming, synchrony, lake characteristics, lake survey

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Contents

Appendix

Papers I-IV

The present thesis is based on the following papers, which will be referred to by the Roman numerals:

I. Göransson, E., Johnson, R.K. & Wilander, A. 2003. Representativity of a mid-lake surface water chemistry sample. (Submitted).

II. Göransson, E. & Johnson R.K. 2003. The importance of ecoregions and catchment characteristics for prediction of pH and total phosphorus. (Submitted).

III. Göransson, E., Bringmark, E., Rapp, L. & Wilander, A. 2003. Modeling the effect of liming on calcium concentration in Swedish lakes. (Manuscript).

IV. Fölster, J., Göransson, E., Johansson, K. & Wilander, A. 2003. Synchronous variation in water chemistry for 80 reference lakes in southern Sweden. (Manuscript).

Lake monitoring and this thesis

Water chemistry has been monitored in long-term monitoring programs in Sweden since the 1960s. The purpose of the national lake monitoring program is to get a comprehensive picture of the status of the 100 000 Swedish lakes with regards to environmental problems such as acidification, eutrophication and heavy metals. The national lake monitoring program covers a broad spatial and long temporal gradient. National lake surveys are performed once every fifth year (sampling of 3 000 to4 000 lakes) to study the spatial variation of lake water chemistry. The temporal variation is covered by approximately 180 national and regional reference lakes, which are sampled three times per year or more. When sampling lakes in the lake monitoring programs, usually a mid-lake surface water sample is taken to represent the water chemistry of the lake. Data from the lake monitoring programs can be found at the homepage of the Department of Environmental Assessment (http://info1.ma.slu.se/db.html; 25-Jan-2003).

Through the implementation of the European Water Framework Directive (European Commission, 2000) the lake monitoring program is under going restructuring. At the same time, methods of interpreting lake monitoring data need to be developed to follow up the environmental quality objectives established by the Swedish Parliament in 1998; "Natural Acidification Only", "Zero Eutrophication", "A Non-Toxic Environment" and "Flourishing Lakes and Streams" (http://www.internat.naturvardsverket.se/documents/objectiv/objectiv.htm). To accomplish all the above, analyzes and evaluations of existing lake monitoring data are necessary. Examples of issues of interest are: What factors control the variation of water chemical metrics in lakes? How do we classify lakes in different lake types with regards to water chemistry? How do we separate human-induced perturbations from natural variation? What is the acidification status of Swedish lakes today? Can limed lakes be classified as acidified? How can we connect the results from the national lake surveys to the results from the temporal reference lakes to improve the interpretation of the status of the Swedish lakes?

Knowledge of indicator variability (*e.g.* total phosphorus and pH), both spatial and temporal, as well as natural and human-induced, is essential for our interpretation of anthropogenic effects. Understanding the spatial variability within and among lakes as well as the temporal variation of lakes is useful for optimizing sampling design. Moreover, this information is needed to improve our awareness of the processes and mechanisms driving indicator metrics.

In this thesis I refer to the issues presented above. I have focused on how spatial variability both within and among lakes affects our interpretation of lake monitoring data (Table 1). In particular, I was interested in assessing the importance of spatial scale, from small (variability within a lake and variability among neighboring lakes) to large (*e.g.* ecoregion and EMEP grid variability), on selected physico-chemical metrics. To improve the interpretation of spatial data, synchronous variation of lakes was also investigated.

Table 1. *An overview of the Papers in this thesis*

	Paper I	**Paper II**	**Paper III**	**Paper IV**
Lake monitoring data	My own sampling of national reference lakes	The 1995 Swedish national lake survey	The 2000 Swedish national lake survey	National and regional reference lakes, 1988-2001
Number of lakes	34	2 765	196 + 255 (totally 451)	80
Geographical distribution of lakes	Whole country	Whole country	Southern and middle part of Sweden	Southern part of Sweden
Scale of the study	Individual lakes and eco-regions	Ecoregions and country	Two EMEP-quadrates (150*150 km)	Southern part of Sweden
Water chemical variables	pH, alkalinity, conductivity, water color, temperature, total P, NO_2+NO_3-N	pH and total P, chemical variables (18), catchment specific variables (28)	Ca, Mg	Indicators of acidification; Ca, SO_4, water color, ANC
Title	*Representativity of a mid-lake surface water chemistry sample*	*The importance of ecoregions and catchment characteristics for prediction of pH and total phosphorus in lakes*	*Modeling the effect of liming on calcium concentration in Swedish lakes*	*Synchronous variation in water chemistry for 80 reference lakes in southern Sweden*

Introduction

Spatial scale

Integrating pattern and scale is a central theme in ecology (Levin, 1992). Ecoregional ecology and landscape ecology are two areas that focus on the interaction between regional patterns and ecological processes at different scales (Turner, 1998; Ormerod, 2000). The importance of a lake's position in the landscape, and especially underlying geology and land use/cover, has provided insight into differences among aquatic systems (*e.g.* Magnuson & Kratz, 2000). Furthermore, the lake trophic classification schemes developed by Thienemann (1925) and Naumann (1932) are examples of the importance of interactions between terrestrial and aquatic systems.

The chemistry of a lake generally reflects the ecological, geological and climatic processes within and around the lake (Gorham, 1961). Moreover, a number of processes influence lake water chemistry at different scales; from small-scale, within-lake variability reflecting in situ processes to large-scale, among-lake variation being primarily determined by catchment geology and hydrology. In particular, geographic position, catchment-scale variability in geology, land use and vegetation cover have provided insight into differences among lake ecosystems (*e.g.* Soranno *et al.*, 1999; Kratz & Frost, 2000). Recent studies have shown that spatial organization and connectivity are important descriptors of lake types. For example, lake chemistry and biology have been shown to be correlated with simply the position of a lake in the landscape (*e.g.* Riera *et al.*, 2000). However, the scale at which we perceive our study objects will, in most cases, influence our interpretation of driving processes and mechanisms. For example, relationships between climate and vegetation that are evident at a broad scale may disappear when finer scales are studied (*e.g.* Wiens, 1989).

Although recent focus has been placed on monitoring whole catchments or ecosystems (Omernik, 1994 and references cited therein) in regional monitoring programs, individual lakes often exhibit marked spatial and temporal variability. Though the spatial-vertical variation generally found within a lake is usually accounted for in sampling, much less attention is given to surficial or horizontal spatial variation. Spatial variation and correlation related to horizontal distances between sampling locations is thoroughly studied within terrestrial systems (Webster & Oliver, 1990). A number of lake studies have focused on the spatial (horizontal) variation associated with phyto- and zooplankton communities (*e.g.* Avois *et al.*, 2000; Lacroix & Lescher-Moutoué, 1995; Pinel-Alloul, 1995; Visman *et al.*, 1994), but much less is known of the horizontal variability associated with lake chemistry. Indeed, spatial patterns in water chemistry are rarely reported, and if mentioned, it is usually done in the context of explaining the spatial variation of plankton (*e.g.* Dickman *et al.*, 1993). In monitoring programs that are temporally extensive (*i.e.* long-term), the horizontal variation in lake water chemistry is important for interpreting long-term changes. Factors such as lake morphometry (*e.g.* depth, shoreline development and volume), distance to shoreline, relative importance of surface and groundwater inputs and biological

activity may separately or combined affect the spatial variation within a lake. In addition, temporally important factors may also affect water chemistry, such as within- and among-year variability in precipitation, temperature, wind speed and direction.

Ecoregions are areas of relatively homogeneous ecological systems, with similar vegetation and climate. It is generally thought that ecoregions with different landscapes have different water quality (*e.g.* Hughes & Larsen, 1988). For example, studies have assessed how well landscape (ecoregion) level classifications can partition physico-chemical and biological variance of lakes (*e.g.* Hawkins *et al.*, 2000). If the transition zone between regions is somewhat diffuse, however, boundary effects might not be evident and differences in biotic assemblages between regions may be difficult to distinguish. Hawkins *et al.* (2000) showed that ecoregions were effective in partitioning variance when distinct differences in geomorphology or strong gradients in physiology and/or climate occurred. Such gradients are found in Sweden; for example, the large-scale regional effect of the *limes Norrlandicus* ecotone (basically a demarcation between deciduous forests in the south and coniferous forests in the north) has been shown to strongly affect different biological communities (*e.g.* littoral invertebrates, Johnson, 2000). Large-scale patterns in lake water chemistry have also been shown to correlate with ecoregion delineations in Sweden (*e.g.* Johnson, 1999). Adopting a spatial approach to partition variance, the European Water Framework Directive proposes the use of ecoregions to classify lake typologies (European Commission, 2000).

Besides naturally occurring variability, aquatic ecosystems often display the effects of a number of human-induced perturbations. For example, lakes in Sweden are affected by eutrophication and acidification, and much of this variance is contagiously distributed, with lakes in the southern parts of the country being more strongly affected than lakes in the north. According to the 1995 national lake survey, between 3.6% (> 50µg P/l) and 5.5% (> 1.5 mg N/l) of 4 113 lakes were classified as very nutrient-rich (Wilander *et al.*, 1998). In the densely populated areas in the south of Sweden with intensive agriculture, 63% of the lakes were classified as very nutrient rich (> 50µg P/l). Furthermore, due to acidification many lakes in Sweden, in some areas as many as 60% of those sampled, are limed or indirectly affected by liming.

Acidification due to human-induced perturbations is mainly caused by acid deposition of sulfur and nitrogen. The level of sulfur and nitrogen which can be tolerated is quantified by the concept of critical load. Critical load is defined as the load below which no harmful effects to the environment occur (UN-ECE, 1994). Large areas of Sweden are sensitive to acidification with low critical loads (Rapp, 2001). At a European level, airborne pollutants are studied by the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP). The pollutants are studied at the scales of the EMEP grid (one EMEP150 grid quadrate = 150*150 km, one EMEP50 grid quadrate = 50*50 km). Critical loads are also modeled at the scale of the EMEP grid size. Since a large proportion of Swedish lakes are affected by liming (20% of the lakes in the national lake survey in 1995 and 27% in 2000), it is important that we can include them when we model critical loads as well as when we

estimate the acidification status of Swedish lakes. Attempts have been made to model the calcium concentration in Swedish lakes affected by liming as if they were unaffected by liming (Henriksen *et al.,* 1990; Wilander *et al.,* 1998). In these studies the relationship between calcium and magnesium (Ca/Mg-ratio) in lakes unaffected by liming was used, assuming that the Ca/Mg-ratio in lakes unaffected by liming was constant. In Henriksen *et al.* (1990) the Ca/Mg-ratio was based on all lakes unaffected by liming with pH below 6.0. In Wilander *et al.* (1998) the Ca/Mg-ratios were based on the lakes unaffected by liming within a county (24 counties in Sweden).

Synchronous variation

An important task in environmental monitoring is to separate natural variation from human-induced perturbations. Reference lakes in monitoring programs are selected to have a minimum influence by such perturbations. If a change occurs in a lake, that lake can be compared to a nearby reference lake to determine if the change is caused by human-induced perturbations or by natural variation. The condition for this comparison is that the variable of interest varies synchronously in the two compared lakes.

Synchronous variation can be expected for lakes influenced by similar conditions, such as climate (*e.g.* precipitation and temperature), deposition and geology (*e.g.* land use and soil chemistry). Synchronous variation of lakes with similar water-chemistry has been shown in several studies (*e.g.* Baines *et al.*, 2000; Pace & Cole, 2002). In a study of ten lakes in Alaska, synchronous variation in lake water chemistry was found for lakes with similar catchment to lake area ratios (Kling *et al.*, 2000). Synchronous variation in lake chemistry can also be expected for lakes that are hydrologically connected (*e.g.* Baines, *et al.,* 2000; George *et al.,* 2000). Finding the most determinant factors (*i.e.* climate, water residence time and water chemical variables) explaining the synchronous variation for lakes would be useful when interpreting results from large contemporary lake surveys.

The ecological status of the whole population of lakes is often investigated in large contemporary surveys. The outcome of such survey is highly dependent on the conditions that prevailed when the survey was performed. For example, the national lake survey in 2000 was performed during extremely high flow conditions, which had an important impact on the water chemistry (Wilander *et al.*, In prep.). Time series from reference lakes are then useful for interpreting data from the national lake survey. However, in order to make the national lake survey valid for average conditions, better knowledge of the rate of synchrony between lakes is needed. We also need to investigate the determinant factors explaining the synchronous variation in lake water chemistry.

Objectives

The objective of this thesis was to study the variability of selected physico-chemical metrics at different spatial scales, from small-scale (variability within a lake and among neighboring lakes) to large-scale (ecoregions, EMEP-grid and parts of the country). I was interested in assessing how different spatial scales would affect our interpretation of lake monitoring data. To further develop the interpretation of spatial data, I also investigated the synchronous variation in lake water chemistry. All studies were based on data from the Swedish national lake monitoring programs (Table 1).

Paper I

The purpose of this study is to address the question if a mid-lake sample can be considered representative of lake water chemistry by studying the horizontal variation of surface water chemistry. My hypotheses are that: (i) where a sample is taken within a lake will affect its representativity; and (ii) a mid-lake sample can be considered as representative of average surface water chemistry.

Paper II

The purpose of this study is to investigate the importance of ecoregions and catchment characteristics for prediction of pH and total phosphorus (TP). Predictive models are evaluated both on ecoregional and whole-country scales. The models are based on catchment-specific variables such as geographic position, catchment land use/cover, and deposition. The hypotheses of the study are that: (i) models based solely on catchment-specific variables can be used to predict pH and TP; and (ii) ecoregion based models will have higher predictive power than whole-country models.

Paper III

It is important to include lakes that are affected by liming in the modeling of critical loads as well as in the estimation of the acidification status. However, lakes affected by liming are difficult to evaluate. The objective of this study is to model the non-marine calcium (Ca^*) concentration in lakes affected by liming (AL-lakes) as if the lakes were not affected by liming. My hypotheses are that: (i) I can model the Ca^* concentration in EMEP lake-populations using the Ca^* and Mg^* concentrations in neighboring lakes unaffected by liming (UL-lakes); and (ii) there is a spatial dependence for Ca^* and Mg^* concentrations in lakes, *i.e.* the Ca^* and Mg^* concentrations in lakes situated close to each other are more similar than in lakes at situated at further distances. For modeling the Ca^* concentration I assume that the Ca^*/Mg^*-ratio for UL-lakes is constant. I further assume that the Mg^* concentration is unaffected by liming. The Ca^* concentration is modeled for lakes in two 150*150 km quadrates, the same spatial scale as one EMEP150 quadrate. The quadrates represent different parts of Sweden with varying loads of acid deposition as well as different weathering rate of Ca^* and Mg^*

(http://www.smhi.se/sgn0102/n0205/nvv/1999/chemdata.htm; 25-Jan-2003 and http://www-markinfo.slu.se/sve/kem/vittring.html; 25-Jan-2003).

Paper IV

The objective of this study is to determine the rate of synchrony between 80 reference lakes in the southern half of Sweden and what factors that control the synchronous variation between lakes. The synchronous variation, 1988 – 2001, is determined for all lake-pairs. The variables studied are indicators of acidification; non-marine calcium (Ca^*), non-marine sulfate (SO_4^*), water color, and Acid Neutralization Capacity (ANC). My hypotheses are that: (i) lakes influenced by similar conditions (*e.g.* climate, deposition and geology) have a high rate of synchronous variation; and (ii) the rate of synchrony for lake-pairs is dependent on the distance between the lakes and how similar the lake and catchment characteristics are (*e.g.* water residence time and different water chemical variables).

Materials and methods

Paper I

The lakes in this study are a part of the Swedish national lake monitoring program. These lakes are sampled annually for physico-chemical parameters. The total number of reference lakes is about 100, and 34 of these were included in this study. The lakes were selected to cover a wide range of lake types. This was done by classifying the lakes according to: (i) geographic region (3 major ecoregions), (ii) water residence time (2 categories, residence time ≤ 1 year or residence time > 1 year), and (iii) water depth (2 categories, mean depth ≤ 3.5 m or > 3.5 m) (Fig. 1a). Since most reference lakes are situated in the southern part of Sweden, which is more densely populated, 5 lakes were selected for each classified group in the southern mixed forest region. In contrast, due to fewer monitoring lakes situated in the north, it was difficult to fill all the groups in the arctic/alpine region and the northern coniferous forest region (Table 2).

Ten samples were taken from each lake from quadrates randomly distributed to cover the lake surface area. Each lake was covered with a grid matrix and the quadrates were numbered, starting in the north end of each lake and moving southward in west to east direction. The total number of quadrates per lake was divided by 10 to determine the interval between sampling sites. The first sampling quadrate was randomly placed in the first interval. The quadrate closest to the center of the lake, according to the lake surface area and where the maximum depth of the lake was located, was chosen as the mid-lake quadrate. Surficial water samples (0.5 meters' depth) were collected from each quadrate during each season. Two sampling seasons, representing high (late summer or August) and low (October) biological activity, were studied. All 34 lakes were sampled in August and 32 in October, 1999.

Eight chemistry variables were studied: pH, alkalinity (meq/l), total phosphorus (µg/l), NO_2+NO_3-N (µg/l, hereafter referred to as inorganic nitrogen), conductivity (mS/m 25 °C) and absorbance (420nm) of filtered water (*i.e.* water color), absorbance (420nm) of unfiltered water, and water temperature (°C).

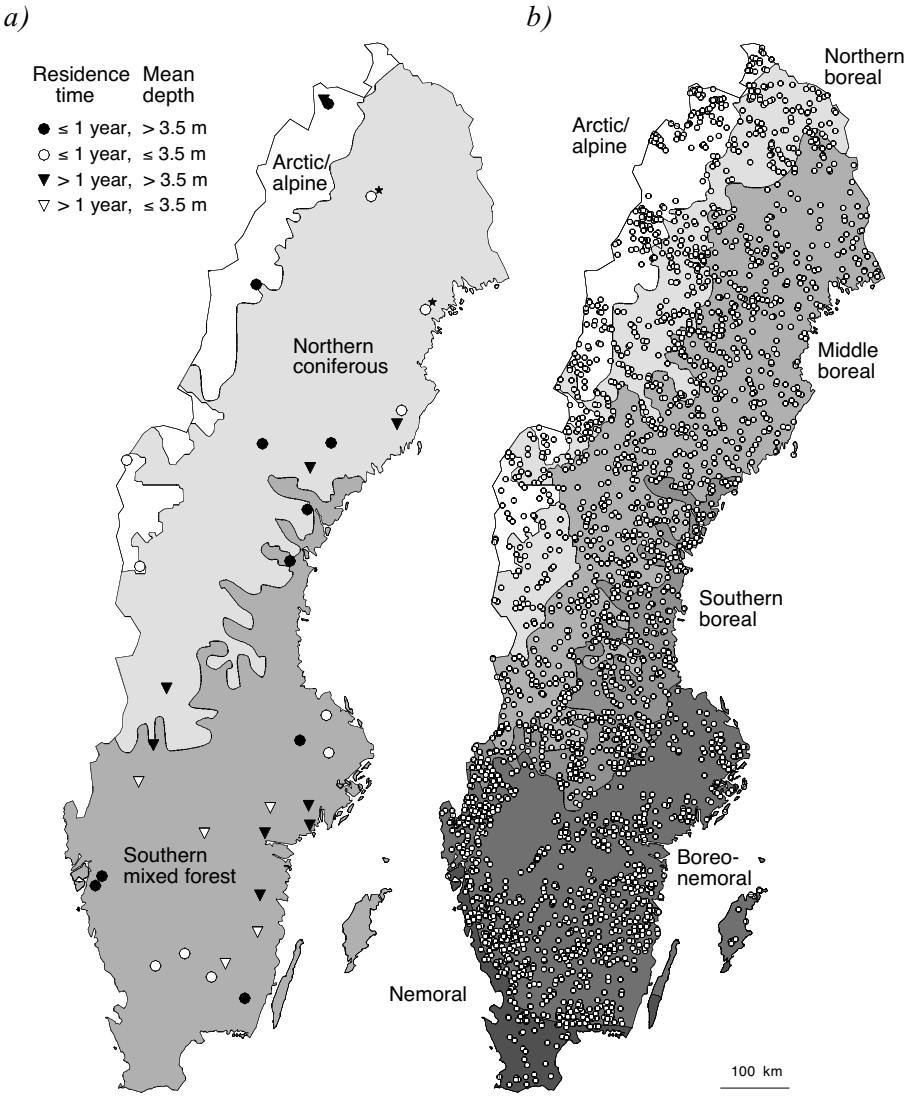

Fig. 1. a) Classification of the 34 national reference lakes according to three ecoregions, water residence time, and mean water depth. The lakes were sampled in August and October 1999. ★ two lakes only sampled in August. *b)* Map showing the six major ecoregions of Sweden and the location of the 2 765 lakes sampled for physico-chemical and catchment-specific (GCD) varibles in the 1995 national lake survey.

Large-scale spatial differences in water chemistry were analyzed by a three-factor unbalanced analysis of variance (ANOVA), with ecoregions (three ecoregions), water residence time (two categories), and mean depth (two categories) as fixed factors. The interaction terms of these three factors were also determined. A Tukey test was carried out to investigate significant differences among the three factors and their interaction terms. A two sample *t*-test was used to determine if the mid-lake sample differed from the nine remaining samples. This was performed on each lake and variable (significant level $\alpha = 0.05$). A jack-knife technique was used to determine if the distance to the center of the lake would affect the representativity of the sample. This was done by sequentially comparing each sample with the remaining nine samples using a two sample *t*-test and Bonferroni correction of α error to a significant level $\alpha = 0.005$.

Table 2. *Number of lakes in each classified group*

Residence time	**≤ 1 year**		**> 1 year**		
Mean depth	**≤ 3.5 m**	**> 3.5 m**	**≤ 3.5 m**	**> 3.5 m**	**Total**
Arctic/alpine region	2	2	-	1	5
Northern coniferous region	3*	3	-	3	9
Southern mixed forest region	5	5	5	5	20
Total	10	10	5	9	34

* Only one lake was sampled in October

Paper II

A Swedish national lake survey was performed in the autumn of 1995. A mid-lake sample was taken in 3 025 selected lakes in Sweden (Wilander *et al.*, 1998). The lakes were randomly selected with some predefined criteria to cover different lake types and to obtain a spatial distribution that covered the whole country (summarized in Henriksen *et al.*, 1996). The samples were collected in the autumn during the mixing period to obtain a representative whole-lake sample. The physico-chemical variables analyzed can be grouped in four categories: indicators of acidity, nutrients, organic material, and major constituents. The survey also included variables of geographic position, altitude, catchment area, and lake surface area. Some 2 700 lakes were also classified according to land use/cover (see Table 2 in Paper II). In total, 2 765 lakes were used in this study. For pH modeling, lakes affected by liming were removed, resulting in 2 046 lakes in the pH models.

All lakes were classified according to ecoregion (Fig. 1b). Partial least squares regression (PLS) was used to evaluate spatial patterns in lake surface water chemistry and to assess the predictive power of models for pH and TP using descriptive and chemical variables (Esbensen *et al.*, 1996; Geladi & Kowalski, 1986). PLS is a multivariate regression method related to principal component analyses. One advantage with PLS is that it works well with intercorrelated data. PLS was used to calibrate and validate predictive models for pH and TP. Two types of models were constructed: (i) models using geographic (G), catchment area classification (C), and deposition (D) variables, respectively, as independent

variables (GCD models); and (ii) models using both GCD and chemical variables and independent variables (GCD+CHEM models). Models were developed for the six ecoregions and for the whole country. Root mean square error of prediction (RMSEP) together with r^2 (explained variance) were used to compare the various models.

Paper I and Paper II – Ecoregions

In Paper I, the ecoregions used correspond closely to those recommended by the European Water Framework Directive (European Commission, 2000). These three regions were originally based on macroinvertebrate distribution studies made in the 1960s (*e.g.* Illies, 1966). The three ecoregions are composed of the ecoregions described below. Hereafter the three regions will be referred to as the arctic/alpine region, the northern coniferous region and the southern mixed forest region (Fig. 1a and Table 2).

In Paper II, data was divided into the six major ecoregions of Sweden using the boundaries given by the Nordic Council of Ministers (1984), but modified according to Gustafsson and Ahlén (1996). The ecoregions range from the nemoral region in the south to the arctic/alpine region in the north (Fig. 1b). The nemoral region is characterized by deciduous forests, mean annual temperature greater than 6 °C, and a relatively long growth period (180 - 210 days) (Clason & Granström, 1992). In contrast, the arctic/alpine region is characterized by relatively low mean annual temperature (< 2°C) and short growth period (< 100 - 120 days). The number of lakes in each ecoregion were (the numbers within parenthesis are the number after removing the lakes affected by liming): 238 (238) in the arctic/alpine region; 191 (188) in the northern boreal region; 978 (843) in the middle boreal region; 389 (206) in the southern boreal region; 912 (529) in the boreonemoral region; and 57 (42) in the nemoral region.

Paper III

A Swedish national lake survey was performed in the autumn of 2000. A mid-lake sample was taken in 3 464 selected lakes in Sweden (Wilander *et al.*, In prep.). The lakes were randomly selected with some predefined criteria to cover different lake types and to obtain a spatial distribution that covered the whole country (summarized in Henriksen *et al.*, 1996). The samples were collected in the autumn during the mixing period to obtain a representative whole-lake sample.

I chose to study the lakes within two 150*150 km quadrates (Fig. 2a), which is the same size as one EMEP150 grid. The quadrates were located in the middle part of Sweden (the Dalarna-quadrate) and in the southern part of Sweden (the Småland-quadrate)(Fig. 2a). The criteria for selecting these quadrates were: (i) they should consist of a large number of AL-lakes and a large number of UL-lakes (Table 3); and (ii) they should differ in acid deposition load. The acid deposition load was higher in the Småland-quadrate than in the Dalarna-quadrate.

The Ca and Mg concentrations were adjusted for marine deposition of Ca and Mg ions using the Cl concentration (Umweltbundesamt, 1996). Non-marine

components of ions are denoted by *. Descriptive statistics were calculated for the two quadrates. For each quadrate, statistics for UL- and AL-lakes were calculated separately. For UL-lakes statistics were also calculated for lakes with Ca^* concentration lower than or equal to 0.5 meq/l.

To find out whether Ca^* and Mg^* in lakes were spatially dependent, I analyzed the data in the two quadrates geostatistically. With geostatistical analyzes it is possible to analyze the spatial variation in a data set (Webster & Oliver, 1990; Oliver & Webster, 1991). I used variogram (isotropic) modeling with GS+ (Robertson, 2000). UL- and AL-lakes were analyzed in separate variograms.

A variogram describes if there is a relationship between distance and variance (semi-variance). When a variable is spatially dependent, the semi-variance increases with the distance. This means that sites at close distances are more similar than lakes at further distances. The distance where the semi-variance reaches maximum is the distance where the sites are spatially independent. If the semi-variance does not increase with distance there is no spatial dependence in the data. Such a variogram is called a pure nugget variogram.

The Ca^* concentration model was based on the assumption that the Ca^*/Mg^*-ratio was constant for UL-lakes. I also assumed that the Mg^* concentration in AL-lakes was unaffected by liming.

I modeled the Ca^* concentration in a lake using the mean Ca^*/Mg^*-ratio for the three and the seven closest neighboring UL-lakes. The Ca^* concentration was modeled according to formula 1.

$$[Ca]_{\text{modeled}} = \frac{\sum_{i=1}^{n}\left(\frac{[Ca]}{[Mg]}\right)}{n} \times [Mg]_{\text{modeled lake}} \qquad (1)$$

where $[Ca]_{\text{modeled}}$, is the modeled Ca concentration (meq/l), $[Mg]_{\text{modeled lake}}$, is the Mg concentration in the modeled lake (meq/l), and n is the three or seven closest lakes.

If any of the three or seven closest neighboring lakes were located outside the quadrates, they were also included in the modeling. The lakes that Ca^* was modeled for were all situated inside the two quadrates. I modeled Ca^* concentrations for UL-lakes as well as for AL-lakes. I validated the models by comparing modeled Ca^* with measured Ca^* in UL-lakes. For UL-lakes, I only modeled lakes with measured Ca^* concentration less than or equal to 0.5 meq/l. I did not have any concentration limit for neighboring lakes included in the Ca^*/Mg^*-ratio. For the Dalarna-quadrate I also modeled Ca^* based on the UL-lakes within a 50-km radius from the modeled lake.

Modeled Ca^* concentration and measured Ca^* concentration were also compared for AL-lakes. For the modeled Ca^* concentration to be valid it must be less than the measured Ca^* concentration.

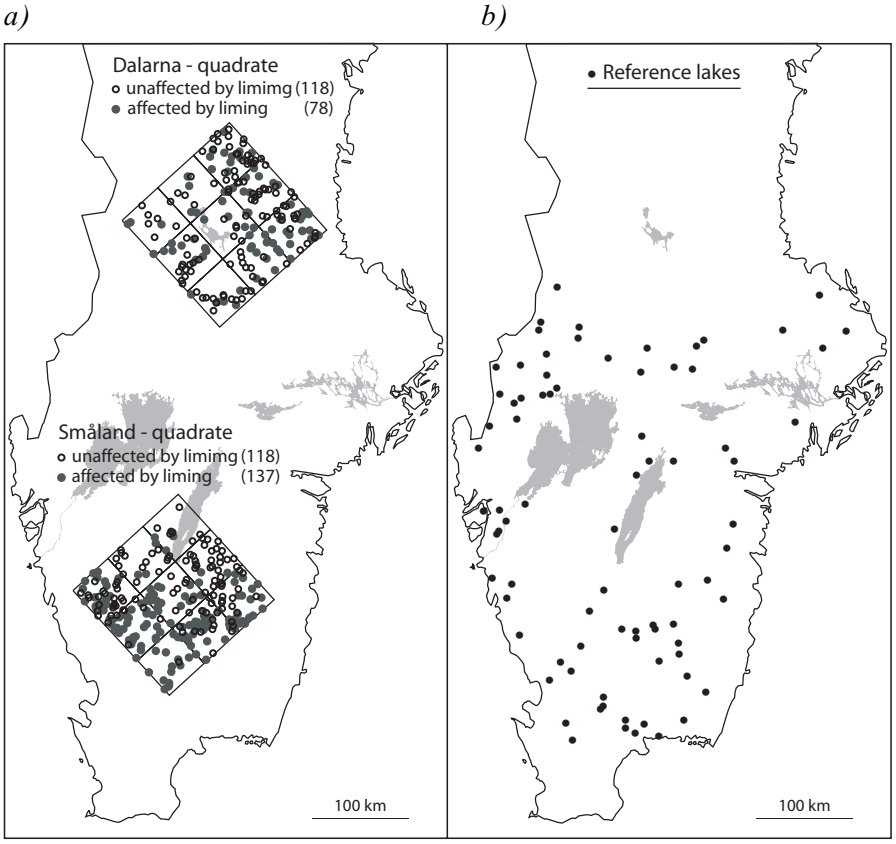

Fig. 2. a) The location of the two selected quadrates. Each quadrate is 150*150 km (the nine smaller quadrates are EMEP50-quadrates). The middle part of Sweden is represented by the Dalarna-quadrate and the southern part by the Småland-quadrate. In each quadrate, there are a large number of lakes unaffected by liming and affected by liming, respectively. *b)* The location of 80 national and regional reference lakes in the southern part of Sweden.

Table 3. *Number of lakes affected and unaffected by liming in each quadrate*

Sampling quadrates	Affected by liming	Unaffected by liming	Total number of lakes
Dalarna	78	118	196
Småland	137	118	255

Paper IV

This study was based on 80 national and regional reference lakes in the southern part of Sweden (Fig. 2b). Sampling of reference lakes started in 1983. As the monitoring program mainly focused on the acidification problem, most reference

lakes were located in the southern part of Sweden. Since then, monitoring of national and regional reference lakes has been complemented to cover the whole country. At present, there are 182 national and regional reference lakes sampled in Sweden. However, most reference lakes are still located in the southern part of Sweden.

Most national and regional reference lakes are sampled three to four times per year and some lakes even more frequently. I chose to study the autumn sample as the lakes are assumed to be mixed when the autumn sample is taken. The national lake survey is also performed in the autumn. Due to logistical reasons, many reference lakes in the northern part of Sweden are not sampled in the autumn. As most reference lakes were located in the southern part of Sweden, together with the fact that all reference lakes in the northern part of Sweden did not have autumn samples, I chose to study reference lakes in the southern part of Sweden. All national and regional reference lakes selected for this study were sampled during the period 1988 to 2000. Of the 80 selected lakes more than 80% were sampled in October, the rest were sampled in November, September or December.

The water chemical variables studied were calcium (Ca^*), sulfate (SO_4^*), water color (AbsF), and Acid Neutralizing Capacity (ANC). Ca^* and SO_4^* were both adjusted for marine deposition of Ca and SO_4 ions (Umweltbundesamt, 1996). Water color was analyzed as absorbance on a filtered sample at 420 nm in a 5 cm cuvett (AbsF).

The synchronous variation of the 14-year long time series was calculated for each lake-pair (3 160 lake-pairs) and for each of the water chemical variables, respectively. The method used for calculating the synchronous variation was the Pearson product moment correlation coefficient (r). The calculations were made with an Excel-macro in Visual Basic.

The distance between the lakes in each lake-pair was calculated. The distance was compared to the synchronous variation for each lake-pair by linear regression. To further investigate if the synchronous variation was connected to how similar the lake and catchment characteristics were for each lake-pair, I calculated the relative difference D_x for various lake and catchment characteristics (formula 2).

$$D_x = \frac{|x_a - x_b|}{x_a + x_b} \qquad (2)$$

where x_a and x_b are the characteristics for lakes a and b. D_x was calculated for 21 variables: lake area, catchment area, the ratio between lake and catchment area, water residence time, percent mire in the catchment, percent forest in the catchment, percent open land in the catchment, percent lake area in the catchment, Ca^*, Mg^*, Na^*, K^*, sum of non-marine base cations, SO_4^*, Cl, alkalinity, NO_3, TP, AbsF, total organic carbon and Si. The synchronous variation and the relative difference were compared by linear regression.

Results and discussion

Paper I

Comparison of the mid-lake sample with the 9 remaining samples for all lakes and variables showed that from 254 (August) and 253 (October) possible combinations, only 10 differed in August and 9 in October. Jack-knifing showed that the location of a sample was important for its representativity of average surface water chemistry. For example, samples taken in the center of the lakes had few differences in water chemistry, indicating that these samples are more representative of average conditions. In contrast, samples taken furthest from the center of the lakes often differed markedly from the other samples (Table 4) (*e.g.* samples taken in the first quadrate). This was especially evident for samples taken close to lake inlets. To further investigate this, I also determined if the mid-lake sample could be considered more representative of lake water chemistry than simply randomly selecting a site. The test was performed in the same way as for the mid-lake sample, the only difference was that the sample was randomly selected from the ten samples for each lake. Of the variables tested, 24 differed significantly in August and 23 in October, compared to 10 and 9, respectively, for the mid-lake sample. Although comparison of only one random sample can not be considered a robust measure of sample representativity, these findings indicate nonetheless that a mid-lake sample is more representative of average lake conditions.

Grouping lakes by ecoregion, residence time and mean depth had a significant effect on a number of variables (except pH, alkalinity and inorganic nitrogen). Ecoregion seemed to be the most important factor describing lake chemistry. Not surprisingly, temperature in August varied significantly for all regions, indicating the large climate gradient that was part of this study. Variables sensitive to biological activity were also influenced by the climate gradient. For example, absorbance of filtered water and total phosphorus also varied with ecoregion. Residence time was also found to be a more important explanatory variable in August than October. This finding might indicate the greater spatial variability expected in late summer (when lakes are relatively hydrologically stable) than autumn (when lakes are turning over). Indeed, several of the variables that varied significantly when the mid-lake samples were compared with the other samples also varied somewhat with season. For example, nutrients seemed to vary more in August than the other variables, whereas variables indicative of weathering and hydrology (*e.g.* alkalinity and conductivity) varied more in October.

Some of the variation within a lake may be attributed to analytical or laboratory-induced variance. For variables exhibiting small, within-lake variability, such as conductivity, this is especially important. I compared the coefficient of variation (CV) of the 10 samples in each lake for each chemical variable (CV_{10}) with the CV of the analytical variation (CV_{av}). I found that CV_{av} was greater than CV_{10} in 2 of the 10 differences noted in August and 6 of the 9 differences in October. If CV_{av} was greater than CV_{10} for the variables tested, I cannot conclude that the mid-lake sample is significantly different for those lakes.

Table 4. *To determine if the distance to the central part of the lake would affect the sample representativity a 2-sample t-test that sequentially compared each sample quadrate with the remaining 9 quadrates by a jack-knifing technique was performed. Each column represents the sample quadrate that was compared to the remaining 9 sample quadrates. The values given show the number of significant differences. P-values were adjusted by Bonferroni corrections ($\alpha \leq 0.005$)*

	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
AUGUST										
pH	9	2					1			1
Alkalinity	2		1	1	1		3			
Conductivity	2						1			
Abs. unfiltered								1		1
Abs. filtered	7		2							1
Total phosphorus										
NO_2+NO_3-N	1				1			1		1
Temperature	1					1			1	1
Sum	**22**	**2**	**3**	**1**	**2**	**1**	**5**	**2**	**1**	**5**
OCTOBER										
pH	3									2
Alkalinity	3		3	2		1			2	1
Conductivity	2			1		1		1	1	2
Abs. unfiltered	2		1	1				1		1
Abs. filtered	6	1		1						
Total phosphorus	1	1			1			1		
NO_2+NO_3-N	2	1								3
Temperature	4		1	1						
Sum	**23**	**3**	**5**	**6**	**1**	**2**	**0**	**3**	**3**	**9**

Paper II

The hypothesis was that ecoregional based models would have higher predictive power (high r^2) and lower error (RMSEP) than the whole country models. This was based on the principle that ecoregions, as integrators of regional patterns in climate, topography, land use, and natural vegetation, would partition part of the natural variability. This seemed to be true for the southern part of Sweden and especially for the nemoral region. Total phosphorus (TP) and pH models developed for the nemoral region had higher predictive power than the model for the whole country and the other ecoregional models (Fig. 3). For the pH and TP nemoral models, GCD variables explained 90% and 57% of the variation, respectively. The total variation within a region seemed to affect the predictive power. For example, total variation within the arctic/alpine region was low, resulting in low predictive power, whereas the total variation within the nemoral region was high, resulting in high predictive power. Overall for both pH and TP, the predictive power of the ecoregional models based on GCD variables was slightly better or about the same as the predictive power for the whole-country models. The prediction error was usually about the same size or somewhat lower for the GCD models based on ecoregions than the whole-country model. An exception was the TP nemoral model, which had a higher prediction error than the whole-country model. One reason might be that there were few lakes in the nemoral region compared to the other regions and the whole country. Some of the pH models seemed to be non-linear across the pH-gradient, with a breakpoint at

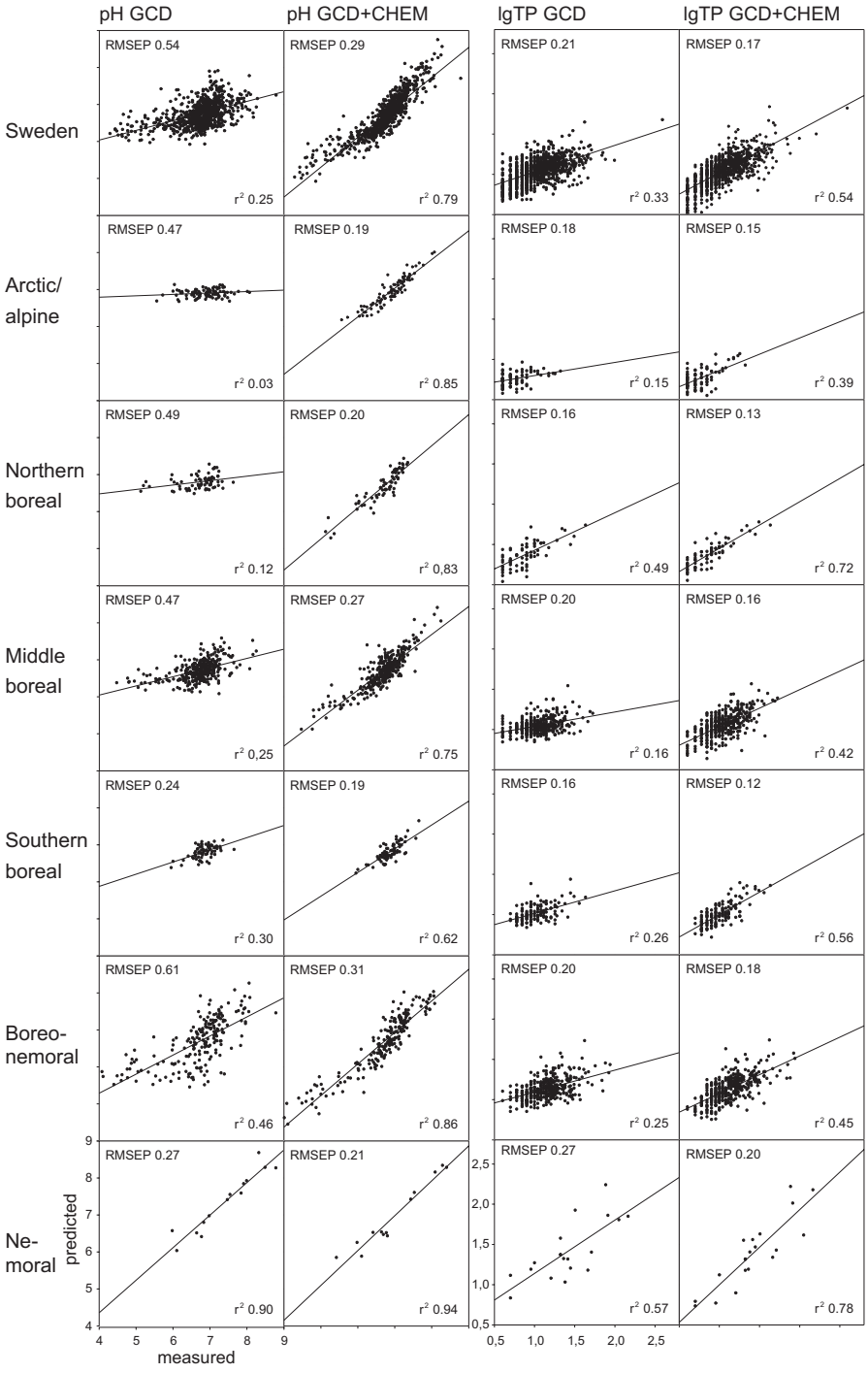

Fig. 3. Predicted vs. measured values for the pH and the $\log_{10}$TP models' validation data sets. The explained variance (r^2) and the predicted error values (RMSEP) are given for the whole-country models (Sweden) as well as for the ecoregion-based models.

approximately pH 6. This implies that it might be better to model pH above and below pH 6 instead of across whole interval. Preliminary studies for the middle boreal region supports this conjecture.

Catchment and lake surface area were important descriptive variables for both pH and TP (GCD models). Other variables such as land use (arable, forest), cover (mire and deciduous forest) and soil type (till) were important for the pH GCD models. Classifications of forest and arable land explained a large part of the variation in pH and TP within the nemoral region. Other important descriptors for the TP GCD models were altitude, and the amount of mires and forests within the catchment (especially in the northern parts of the country).

As expected, adding the chemical variables improved the predictive power of the models considerably. The most obvious example was the arctic/alpine pH model. In this model the explained variance increased from 3% to 85% when the chemical variables were included. The most important predictor for pH was calcium (Ca), whereas the most important for TP were water color (absorbance) and in some cases TOC and the nutrients NH_4-N and total nitrogen (TN). Interestingly, these chemical variables (Ca, water color, TOC, NH_4-N, and TN) all reflect conditions in the catchment.

Paper III

In many cases it is striking how well the results for the two quadrates Dalarna and Småland match. For UL-lakes, Ca^* and Mg^* were spatially dependent as shown by the variograms (Fig. 4). Accordingly, lakes situated close to each other were more similar than lakes at further distances with regard to Ca^* and Mg^* concentrations. For AL-lakes, however, only Mg^* showed spatial dependency, whereas the variograms for Ca^* were almost pure nugget ones. This means that I could not find any spatial dependence in the data for Ca^* concentration in AL-lakes. Earlier studies have shown that lack of spatial dependence within a data set can be caused by human-induced perturbations (Bringmark & Bringmark, 1995; Clarholm & Bringmark, 2003). Human-induced perturbations can disturb the natural variation causing a random variation in the data set. In this study the human-induced perturbation would be liming. Also the fact that Mg^* was spatially dependent for AL-lakes supports my assumption that the Mg^* concentration is not affected by liming. The flatter Mg^* variograms for AL-lakes compared to UL-lakes is due to the lower Mg^* variation among the former lakes (see Table 2 in Paper III).

The Ca^* and Mg^* variograms for UL-lakes in the Dalarna-quadrate reached a maximum semi-variance at the distance of approximately 50 km. The corresponding Mg^* variogram for AL-lakes also reached a maximum semi-variance at approximately 50 km. This means that Ca^* and Mg^* concentrations in lakes within a radius of 50 km are spatially dependent, while the Ca^* and Mg^* concentration in lakes at distances further than 50 km from each other are spatially independent. The distance 50 km is especially interesting since an EMEP50 grid is 50*50 km. All neighboring lakes included in the modeling of Ca^* in the Dalarna-quadrate were located within 50 km from the modeled lake. The farthest distance was 37.6 km. For the variograms that showed spatial dependence for the

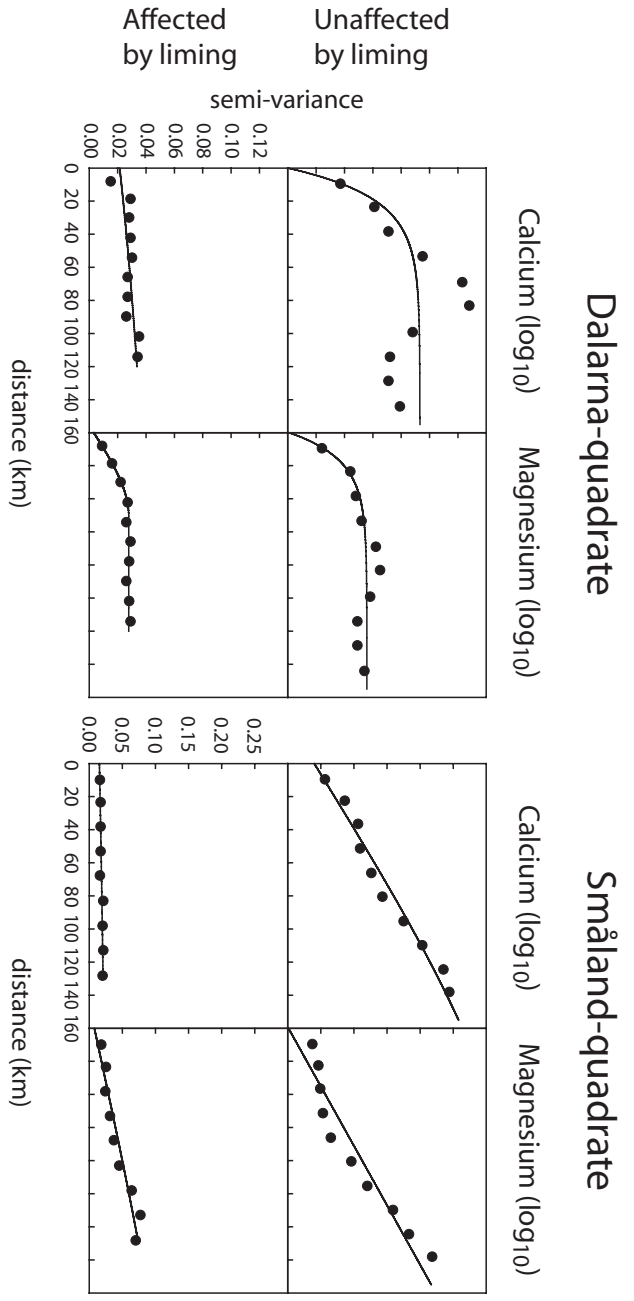

Fig. 4. The modeled variograms for the Dalarna and the Småland-quadrates. For lakes unaffected by liming all variograms show spatial dependency. For lakes affected by liming only the Mg variograms show spatial dependency, whereas the variograms for Ca are almost pure nugget ones. For the Dalarna-quadrate, the variograms for Mg and the Ca variogram for lakes unaffected by liming reach a maximum semi-variance at approx. 50km. The Ca and Mg concentrations used are the non-marine components of Ca and Mg. The Ca and Mg values are $\log_{10}$ transformed.

Småland-quadrate, the semi-variance did not reach maximum within the longest distance of the quadrate. This means that all lakes in the Småland-quadrate are spatially dependent. Again, lakes close to each other were more similar regarding the Ca^* and Mg^* concentration than lakes at further distances.

Validation of the modeled Ca^* in UL-lakes showed good results (Fig. 5). The linear regression of Ca^* modeled versus Ca^* measured captured about 60% of the variation (*i.e.* $r^2 > 0.6$). The slopes of the fitted lines were close to 1 and the intercepts close to 0. Modeling Ca^* with the three or the seven closest lakes gave similar results for both the Dalarna-quadrate and the Småland-quadrate.

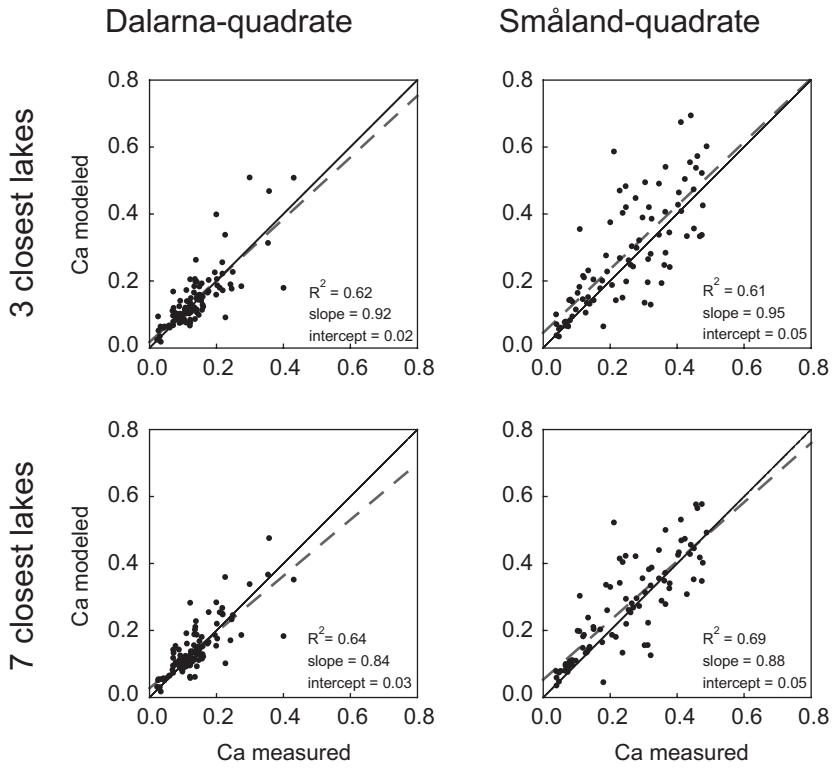

Fig. 5. Modeled Ca vs. measured Ca for both quadrates studied. The modeled lakes are unaffected by liming with measured Ca≤0.5 meq/l. The modeled Ca concentrations are based on the average Ca/Mg-ratios from the three and the seven closest neighboring lakes, respectively. One modeled lake was excluded for the Dalarna-quadrate. The excluded lake had a measured Ca concentration of 0.193 meq/l and modeled Ca concentrations of 0.791 meq/l and 0.568 meq/l for the three and the seven closest lakes, respectively. Excluding the lake did not affect the linear regression appreciably. The Ca and Mg concentrations used are the non-marine components of Ca and Mg.

Comparing the modeled Ca^* with the measured Ca^* for AL-lakes also gave good results. For the Småland-quadrate only 10% (three neighboring lakes) and 9% (seven neighboring lakes) of the modeled lakes had a Ca^* modeled concentration higher than the Ca^* measured concentration. For the Dalarna-quadrate the corresponding results were 36% and 38%, respectively. The larger number of lakes

with modeled Ca^* concentrations higher than the measured Ca^* concentration in the Dalarna- than in the Småland-quadrate is probably due to the higher load of acid deposition in the Småland-quadrate. The mean concentrations of Ca^* and Mg^* in the two quadrates, imply that there is a larger difference between UL- and AL-lakes in the Småland than in the Dalarna-quadrate (see Table 2 in Paper III). Smaller differences between UL- and AL-lakes, makes it more difficult to model Ca^* concentrations in AL-lakes. Some lakes in the Dalarna-quadrate may only be slightly affected by liming and yet still be classified as lime-affected.

Paper IV

The water chemistry in the 80 national and regional reference lakes had a high rate of synchronous variation (Fig. 6). The median r-values were 0.79 for SO_4^* and between 0.51 and 0.57 for AbsF, Ca^*, and ANC. Of the 3 160 lake-pairs, 93% of the r-values were statistically significant for SO_4^* ($p<0.05$), 58% for AbsF, 59% for Ca^*, and 67% for ANC. For AbsF, Ca^*, and ANC, about one fourth of the 3 160 lake-pairs had an r-value larger than 0.71. This means that for one fourth of the lake-pairs, more than 50% of the variation in one of the lakes could be explained by the variation in the other lake. For SO_4^* 70% of the lake-pairs had an r-value larger than 0.71.

The high synchrony for SO_4^* can be explained by the downward trend in SO_4^* during the 1990's, which was caused by reduced SO_4^*-emissions followed by a decline in SO_4^*-deposition. In some lakes, downward trends were also present for Ca^*. However, the trends were weaker than for SO_4^*.

I expected that lakes situated close to each other would have similar conditions regarding *e.g.* climate, deposition and geology, and therefore also be more synchronous than lakes situated far from each other. However, the linear regression of synchrony versus distance, did not show any relationship between synchrony and distance. This means that lakes at short as well as long distances can be synchronous. An example of distant lakes with high synchrony is Lake Lillesjö and Lake Ulvsjö, which are separated by a distance of 402 km and have a rate of synchrony for SO_4^* of 0.93.

When I compared the synchrony with the similarity of various lake and catchment characteristics, I did not find any obvious relationships. The highest r^2-value was 0.13 for the linear regression of Ca^*-synchrony versus the relative difference of Ca^*.

The linear regressions of synchrony versus distance and synchrony versus relative differences in various lake and catchment characteristics were made for all 80 lakes. When I made the same linear regressions but at a smaller scale, *viz*. for 15 lakes in the south-east of Sweden, I found somewhat better relationships. For example, linear regressions of synchrony versus distance for Ca^* and $S0_4^*$ gave the r^2-values 0.49 and 0.28, respectively. The corresponding r-values for synchrony versus relative difference in lake area in the catchment were 0.21 and 0.20. This finding indicates that there might be relationships between the synchronous variation in lake water-chemistry and lake and catchment characteristics in smaller regions.

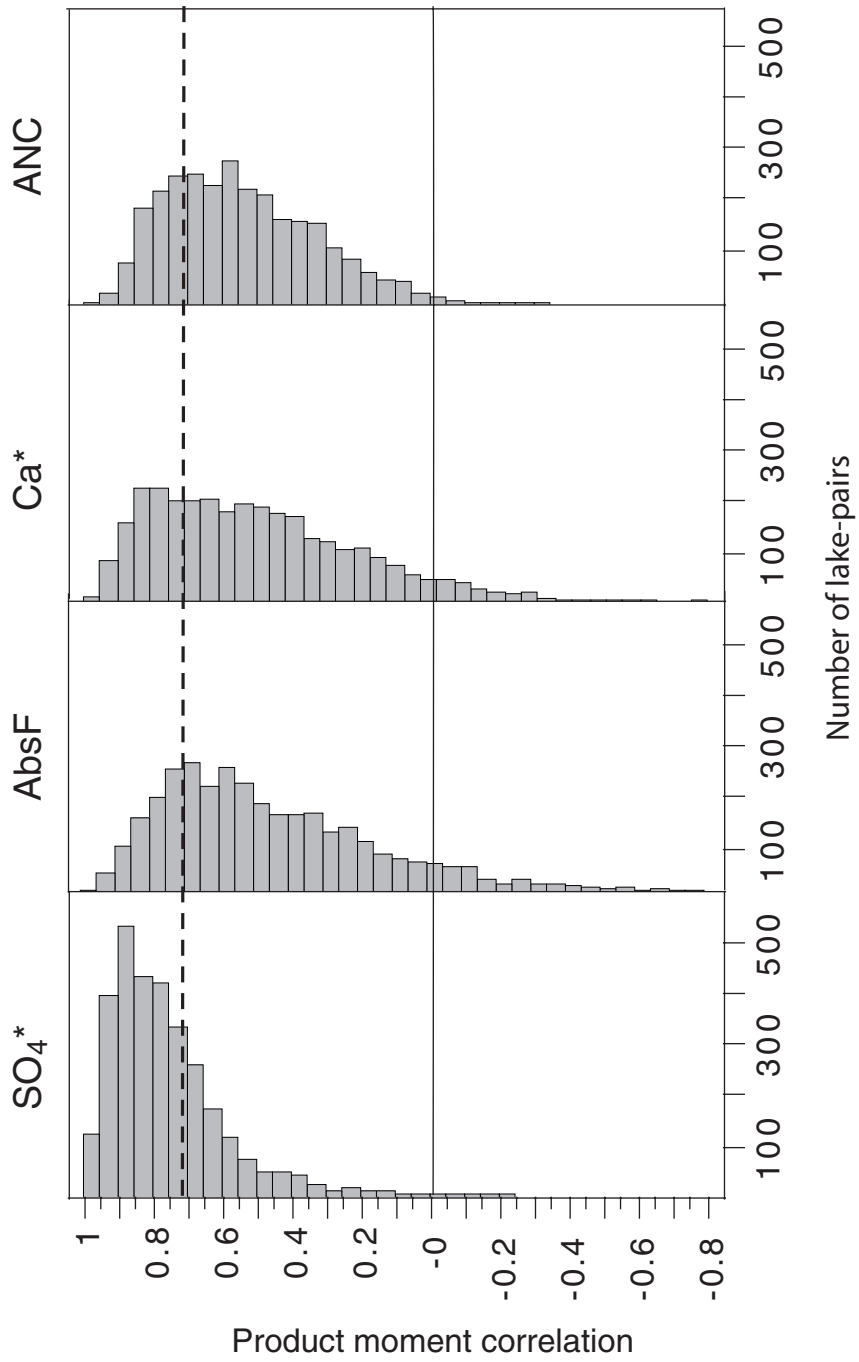

Fig. 6. Synchronous variation of SO_4^*, AbsF, Ca^*, and ANC in 80 reference lakes with 14 year time series. Distribution of synchrony (Pearsons product moment correlation coefficient, r) for 3 166 lake-pairs on the y-axis and number of lake-pairs on the x-axis. Above the dashed line (r=0.71), more than half of the variation in one lake (in the lake-pair) can be explained by the variation in the other lake.

Conclusions and future perspectives

Both large and small spatial scales were found to affect lake water chemistry. Where a sample was taken within a lake was shown to be important for how representative it is in terms of lake surface chemistry (Paper I). In particular, samples taken from quadrates that were situated close to a lake inlet were found to differ. Mid-lake samples, on the other hand, were shown to be representative of average surface water chemistry. However, the finding that water residence time was an important explanatory variable for August water chemistry implies that lakes are more heterogeneous in late summer than autumn. In autumn, many of the lakes were probably circulating, which may partly explain the less heterogeneous conditions measured at this time. However, my results indicate that mid-lake samples taken either during August or October can be considered representative of average conditions and should continue to be used in lake monitoring and assessment programs.

In Paper II, I investigated if models based solely on catchment-specific variables can be used to predict pH and TP and also if ecoregion based models will have higher predictive power than whole-country models. The study showed that pH and TP were best predicted by other in-lake physico-chemical variables, implying that in-lake chemistry was a better predictor than catchment-scale descriptors. However, many of the "better" predictor variables are known to be dependent on characteristics of the catchment. For example, calcium was shown to be a good predictor of lake pH. Calcium is also strongly correlated with catchment geology and soil chemistry. Hence, pH should be correlated with these catchment-scale variables as well. In this study I did not have access to soil or geological information. Future studies should focus on determining the linkages between catchment-scale variability (*e.g.* geology and weathering) and in-lake chemistry.

Ecoregion models were not always better than whole-country models in predicting pH and TP. In particular, models developed for the southern parts of the country performed better. In contrast, when the within-ecoregion variance was low (*e.g.* in the arctic/alpine region) ecoregion models performed poorly. Better ecoregion-based models may be developed by partitioning among-lake variance using additional landscape delineations (*e.g.* geology and highest post-glacial coastline). However, predictive models for some regions may be more difficult to develop. For example, geology in the arctic/alpine region is known to be very variable.

For most lakes, there are incomplete records of water chemistry before the lakes were affected by liming. This study (Paper III) describes, to our knowledge, the only method to model the Ca concentration in AL-lakes as if they were not affected by liming. The Ca/Mg-ratio has been used in earlier studies to model the Ca concentrations in Swedish AL-lakes. The Ca/Mg-ratios in those studies were based on lakes within the whole country (Henriksen *et al.*, 1990) or within a county (Wilander *et al.*, 1998). In this study I have developed this method further by investigating the spatial dependence of Ca^* and Mg^* concentrations. I also modeled the Ca^* concentrations in lakes with individual Ca^*/Mg^*-ratios for each

modeled lake. The results support the hypothesis that I can model the Ca^* concentrations in lakes using the Ca^* and Mg^* concentrations in neighboring lakes, at least within the quadrates of Dalarna and Småland. It should be emphasized that the models are not precise enough to estimate the Ca^* concentration in a single lake. However, the models are good estimators of Ca^* concentrations for EMEP lake-populations and also for estimating critical loads and the acidification status at the EMEP-scale. In the future, it would be interesting to develop models for single lakes; however, more data material of lake and catchment characteristics are needed (*e.g.* geological information and weathering). As there are spatial dependence in the Ca^* and Mg^* concentrations one method to develop the models further is kriging (*e.g.* Bellehumeur *et al.*, 2000).

Lakes seem to have a high rate of synchronous variation, at least for the variables SO_4^*, Ca^*, AbsF and ANC. Even though the 80 lakes in Paper IV were not hydrologically connected they were highly synchronous. At a large-scale, *i.e.* for 80 lakes in the southern part of Sweden, the synchrony of lakes does not seem to be related to distance between the lakes or to any other of the lake characteristics I studied. However, at a smaller scale the study indicates that there might be regions where there is a relationship between synchrony and lake characteristics as well as a relationship between synchrony and the distance between lakes. This finding is interesting for classifying Sweden into water chemical regions. However, to increase understanding of the relationship at a smaller scale, further studies are needed.

If I were a rich woman *(freely from "Fiddler on the roof")*

In this thesis I have studied the variance in lake water chemistry at different spatial scales using different approaches. Understanding the spatial variability within and among lakes is important for optimizing sampling design as well as improving our awareness of the processes and the mechanisms driving indicator metrics. Partition lakes regarding lake water chemistry is an important issue for the interpretation of lake monitoring data, *e.g.* for explaining trends in water chemistry. Within the European Water Framework Directive (European Commission, 2000) characterizing lake-types and partition lakes into lake-types is a central issue. According to the European Water Framework Directive, two of the criteria to classify lakes are ecoregions (three regions) and topography (three elevation above-sea-level classes). Topography, soil type and vegetation are examples of physiographic factors that Gottschalk *et al.* (1979) believe are important for classifying Sweden into hydrological regions. Another important factor for such classification is runoff (Tollan, 1975; Gottschalk *et al.* 1979). Classifying data in ecoregions is a recognized method used in studying biology (*e.g.* littoral invertebrates, Johnson, 2000). However, Paper II showed that classifying lake water chemistry into ecoregions (six ecoregions) only works in parts of the country and also that it is important to have sufficient variance within the ecoregions.

The results in my thesis show that there is a spatial dependence in lake water chemistry as well as a high rate of synchronous variation (Paper III and IV). Both Paper III and IV indicate that water-chemical data should be interpreted at a

regional scale (*e.g.* EMEP-scale, 15 lakes in the south-east of Sweden). The spatial dependence between lakes analyzed in Paper III indicates that there is dependence in lake water chemistry, but that the distance for dependence can vary within the country. A study by Laberge *et al.* (2000) shows that the distance for dependence also can vary between years. The results from the 15 lakes in the south-east of Sweden indicate that the factors controlling the synchronous variation will preferably be found at the regional scale. Further studies of the spatial dependence between lakes as well as the synchronous variation, might lead us to methods to classify the country into regions according to lake water chemistry. However, to interpret the water-chemical effects on the biota, it is important that the classification into regions according to water chemistry can be applied to biological variables as well as to water-chemical variables.

Paper II, III and IV support the fact that further studies of geological parameters are needed to further understand the variation in lake water chemistry. We need better knowledge of the connection between variation in lake water chemistry and *e.g.* weathering, soil chemistry, soil type, distribution of mires and land use. Hydrological parameters also interesting for future studies are *e.g.* precipitation, deposition and runoff. It would be interesting to develop a method to classify the country into regions regarding lake water chemistry together with experts in the fields of geology and hydrology (*e.g.* researchers from the Geological Survey of Sweden and the Swedish Meteorological and Hydrological Institute).

Further research to find the answers to the following questions would be an important and exciting challenge:

- What factors control the spatial dependence of lake water chemistry?
- How does the spatial dependence of lake water chemistry vary within the country?
- What factors control the synchronous variation between lake-pairs?
- Can we use the rate of synchronous variation to classify lakes into regions?
- How do we develop models to predict the Ca concentration in a single lake affected by liming as if it was not affected by liming?

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